

## **HYBRID COATINGS DERIVED FROM BITUMINOUS AND PETROLEUM BASES**

### **BACKGROUND OF THE INVENTION**

The present invention resides generally in the field of tars, tar-based enamels and the components of the tar-based enamels used to form hybrid coatings for metal articles to waterproof and protect the coated surface from corrosion. More specifically, the present invention relates to improved enamel bases derived from a mixture of bituminous and petroleum bases that are used to prepare tar-based enamels providing performances similar to current bituminous or coal tar-based enamels and providing reduced levels of polycyclic aromatic hydrocarbons (PAH's). The present invention further relates to methods for making the improved enamel bases and the tar-based enamels, methods for applying the enamels as well as articles treated with the novel enamels.

As further background, bituminous or coal tar-based enamels derived from a coal tar base, coal and a filler have long set the standard of protection for metal articles such as metal pipes and tanks located in corrosive environments. Recent reductions in coke consumption have significantly reduced the amount of available raw coal tar needed for the preparation of coal tar enamels and related products. The current coal tar shortage is expected to worsen and no commercially attractive alternatives for coal tar enamel are presently available.

Increasing environmental limitations on emissions from manufacturing facilities where coal tar enamels are produced and used and concern for the environment where coated articles are placed, have created an increased sensitivity to the level of PAH's in coal tar products. A paper presented by Mirtchi and Noël at Carbon '94 at Granada, Spain entitled "Polycyclic Aromatic Hydrocarbons in Pitches Used in the Aluminum Industry," described and categorized the PAH content of pitches made from coal tar. Mirtchi and Noël identified 14 PAH materials which the United States Environmental Protection Agency considered to be potentially harmful. Each of the 14 materials was assigned a relative ranking of toxicity based on a standard arbitrary assignment of a factor of 1 to Benzo(a)pyrene or B(a)P. Estimations of the PAH levels in coal tar based material can be made by converting its total PAH content into a B(a)P equivalent which eliminates the necessity of referring to each of the 14 compounds individually.

In light of this background, there is a need for new tar-based enamels having the desirable coating properties of a bituminous or coal tar enamel but requiring less coal tar-based raw materials. Such tar-based enamels would retain the desirable properties of a coal tar enamel, be readily produced in part from more abundant and readily available starting materials and have lower B(a)P equivalents than a corresponding coal tar enamel. The present invention addresses these needs.

### SUMMARY OF THE INVENTION

Applicant has discovered a hybrid tar-based enamel that maintains the desirable properties of a coal tar enamel but requires reduced amounts of raw materials derived from coal tar. Additionally, the new tar-based enamel has substantially reduced levels of PAH's compared to traditional coal tar enamels. Accordingly, one preferred embodiment of the invention provides a tar-based enamel comprising an enamel base and an optional filler. The preferred enamel base is a substantially homogeneous blend of at least one bituminous base, at least one petroleum base and coal. Suitable fillers include, but are not limited to, talc, slate, mica, limestone, silica, kalon and other similarly inert materials. The filler, when used, imparts impact strength to the coating and will generally be about 1 to about 40 weight percent of the tar-based enamel. For applications where increased impact and mechanical strength are not needed, such as for example the interior of a tank, an enamel base having a softening point of at least about 90°C can function as a tar enamel without the addition of a filler.

Another preferred embodiment of the invention provides an enamel base having the desirable properties of a coal tar base while having reduced levels of PAH's. Applicant's preferred enamel base is a substantially homogeneous blend of bases derived from bituminous and petroleum sources and coal that alone or when combined with an appropriate filler provides a tar-based enamel meeting the performance specifications of a coal tar enamel.

Another preferred embodiment of the invention provides for a process for producing applicant's tar-based enamel. One aspect of this process comprises forming a mixture of applicant's novel enamel base derived from a mixture of bituminous and petroleum bases and coal with a filler and heating and stirring the components at a temperature of at least about 225°C until the mixture is uniform.

Another preferred embodiment of the invention provides for a process for producing applicant's enamel base. The process comprises forming a mixture of bituminous and petroleum bases and coal, heating the mixture and removing sufficient distillate to provide an enamel base suitable for mixing with an appropriate filler to produce a tar-base enamel meeting the performance specifications for a coal tar enamel. As the mixture is heated, the coal dissolves and as distillate is removed the base's softening point increases. The relative amounts of bituminous and petroleum bases can be varied according to their availability and/or to provide a desirable PAH content, depending on the application.

Still another preferred embodiment of the invention provides for a coated metal article having a surface coated with a tar-based enamel such as for example a steel pipe or steel tank. A preferred coated article has at least one surface either partially or completely coated with the tar-based enamel. A coated surface can be an interior or an exterior surface of the article.

A coated steel pipe includes a steel pipe having a steel surfaces and a tar-based enamel coating on at least one of the steel surfaces. In this respect the steel pipe having a tar-based enamel coating is substantially waterproofed and protected

from corrosion to a similar degree as a pipe having a coal tar enamel coating. Additionally, applicant's coated pipe having a tar-based enamel coating introduces into its surrounding environment reduced amounts of PAH's compared to a pipe having a traditional coal tar enamel coating. The methods used to coat a pipe surface with a coal tar enamel are suitable for coating a pipe surface with applicant's tar-based enamel and are described below.

A coated steel tank includes a steel tank having a tar-based enamel coating on all or at least a portion of an interior and/or exterior surface. In this respect the steel tank having a tar-based enamel coating is substantially waterproofed and protected from corrosion to a similar degree as a steel tank having a coal tar enamel coating. Similarly, applicant's coated steel tank having a tar-based enamel coating introduces into its surrounding environment reduced amounts of PAH's compared to a steel tank having a coal tar enamel coating. The methods used to coat a steel tank's surface with a coal tar enamel are suitable for coating a steel tank's surface with applicant's tar-based enamel and are described below.

The invention thus provides in its various embodiments improved coated articles, improved tar-based enamel coatings, an improved enamel base, and methods for making the tar-based enamel and the enamel base and methods for applying the tar-based enamel. In addition, the coatings contain reduced quantities of PAH's and substantially meet the performance specifications required for coal tar enamels while utilizing reduced amounts of the less available coal tar. Additional objects, features and advantages of the invention will be apparent from the following description.

### DESCRIPTION OF THE PREFERRED EMBODIMENT

For purposes of promoting an understanding of the principles of the invention, reference will now be made to certain embodiments thereof and specific language will be used to describe the same. It will nevertheless be understood that no limitation of the scope of the invention is thereby intended, such alterations, further modifications, and applications of the principles of the invention as illustrated herein being contemplated as would normally occur to one skilled in the art to which the invention relates.

As indicated above, the present invention provides tar-based enamels capable of meeting the product specifications for coal tar enamels, but requiring reduced amounts of raw materials derived from coal tar. Additionally, the tar-based enamels contain lower levels of PAH's compared to a similar enamel derived solely from coal tar. As a result, the manufacture and use of the tar-based enamels having the necessary performance results in: (a) the current supplies of coal tar being extended and (b) the reduced exposure of workers and the environment to PAH's. PAH levels are considered in terms of B(a)P equivalents as discussed above.

In this regard, the term coal tar is well known in the art, and refers to a liquid condensate that results from the destructive dry distillation or carbonization of coal. Similarly, the terms bituminous base and petroleum base are well known and refer to products resulting from the further processing of raw coal tar and from the rectification of petroleum products, respectively. Examples of bituminous

bases include, but are not limited to, coal tar, coal tar pitch, refined coal tar, roofing tar and related materials. Examples of petroleum bases include, but are not limited to, cat cracked clarified oil (a soft petroleum pitch), slurry oil, decant oil, aromatic petroleum oil and related materials. Some typical properties of two petroleum bases are provided in the table below:

| <b>Physical Property</b>     | <b>Cat Cracked Clarified Oil</b> | <b>Slurry Oil</b> |
|------------------------------|----------------------------------|-------------------|
| Density g/cc @60°F           | Min. 1.15                        | 1.08              |
| Softening Point, Ring & Ball | Min. 41°C                        | -----             |
| Sulfur Mass %                | Max. 1.7                         | Max. 1.0          |
| Distillation                 |                                  |                   |
| C/H Ratio                    | Min. 1.05                        | Min. 0.95         |
| Initial BP                   | 371°C                            | 200°C             |
| 50%                          | 453°C                            | 390°C             |
| Final BP                     | >600°C                           | >500°C            |

Table I

The term enamel base is similarly known in the art and refers to a homogeneous mixture containing a tar base and coal that is heated to dissolve the coal and distilled to raise the base's softening point and lower its penetration.

The term coated article refers to an object such as a metal pipe or tank having at least a portion of one or both surfaces treated with a tar-based enamel to waterproof the treated portion of that surface, to provide protection of the treated surface from corrosion or both.

Preferred enamel bases include homogeneous blends of at least one bituminous base, at least one petroleum base and coal that have been processed to provide an enamel base that upon the addition of a filler provides a tar-based

enamel that meets the performance specifications for a coal tar enamel. Examples of specifications for coal tar enamel include those found in NACE standard RP0399-99 "Plant-Applied, External Coal Tar Enamel Pipe Coating Systems: Application, Performance, and Quality Control" Table A1 Physical Properties of Coal Tar Enamel. Specifications can also be found in AWWA C 203 "Coal-Tar Protective Coatings for Steel Water Pipelines-Enamel and Tape Hot Applied".

Preferred enamel bases will typically have a Ring and Ball (ASTM D36) softening point of at least about 70°C and a penetration of at least about 2 as determined by ASTM D5. The relative amounts of bituminous and petroleum bases can be varied depending on the availability of coal tar and the acceptable level of PAH's in the tar-based enamel prepared from the enamel base. Preferred enamel bases will contain from about 10-75% bituminous base, about 10-75% petroleum base and about 10-30% coal and will typically have a B(a)P equivalent at least about 10% less than a corresponding enamel base derived from only a bituminous base and coal. More preferred enamel bases will contain from about 20-50% bituminous base, about 35-60% petroleum base and about 15-25% coal and typically have a B(a)P equivalent at least about 35% less than a corresponding enamel base derived from only a bituminous base and coal. Enamel bases can be directly converted to tar-based enamels or held for later conversion. Additionally, the enamel bases having a sufficiently high softening point can be utilized directly as a tar-based enamel without the addition of a filler for applications where the coating is generally protected from impact.

For ease of dissolution, it is further preferred that the coal component of the enamel base be in the form of coal dust. Larger particles of coal can be used, but require longer heating periods for full dissolution.

To form the enamel base in accordance with this invention, the blend of ingredients can be charged to a still, for instance a batch or continuous still, and with agitation and heating conveniently distilled to provide the desired homogeneous enamel base. Preferred enamel bases are formed by distilling the blend until a maximum liquid temperature of from about 300 to 350°C is reached, when the distillation is conducted at atmospheric or reduced pressures. Additional periods of heating without further distillation can be used to more fully dissolve the coal or to maintain the base as a fluid ready for testing and/or further processing including additional distillation or combination with filler to provide tar-based enamels.

Preferred tar-based enamels are an intimate mixture of applicant's preferred enamel base described above and a filler that are fluid at elevated temperatures. A variety of materials having a particulate form that are generally inert under the tar-based enamel's processing conditions can function as fillers. Preferred fillers include, but are not limited to talc, slate, mica, limestone, silica and kalon.

The preferred tar-based enamel's properties primarily depend on the nature of the enamel base and amount of filler used. Generally enamel bases having increased amounts of coal provide tar-based enamels having increased softening points. Similarly, enamel bases having increased amounts of bases derived from coal or petroleum sources generally provide tar-based enamels having higher

penetrations. Filler levels are adjusted to cause the tar-based enamels to have properties that correspond to a desired product specification. For applications in which the tar-based enamel is subjected to minimal contacts that could cause abrasion or disruption of the coating, filler levels as low as about 1 weight percent have been found to perform satisfactorily. Preferred tar-based enamels contain from about 10-40 weight percent filler and more preferred tar-based enamels contain from about 20-35 weight percent filler. Generally, a greater reduction in the B(a)P equivalents for a tar-based enamel can be obtained by utilizing an enamel base having increased amounts of petroleum bases compared to coal tar bases. The B(a)P equivalents in applicant's preferred tar-based enamels are reduced at least about 20% relative to a coal tar enamel derived solely from bituminous materials and contain from about 20-40 weight percent petroleum base. The B(a)P equivalents in applicant's more preferred tar-based enamels are reduced at least about 40% relative to a coal tar enamel derived solely from bituminous materials and contain from about 40-60 weight percent petroleum base.

Although the product specifications for enamel coatings based on coal tar vary, the preferred tar-based enamels will have Ring and Ball (ASTM D36) softening points of at least about 90°C and penetration determined by the method of ASTM D5 of at least about 2. More preferred tar-based enamels will have softening points of at least about 100°C and penetrations of at least about 5. The performance specification for a coal tar enamel depends on the application for which the enamel is intended. One specification for typical grades of coal tar

enamel is provided in Table II. Other performance specifications include AWWA Physical Properties for Coal Tar Enamel.

| Physical Property and Test                        | Type 1<br>Standard<br>Fully Plasticized |                |  | Type 2<br>Special, Fully Plasticized |                |  | Type 3<br>High-Temperature<br>Fully Plasticized |                |  | Type 4<br>High-Temperature<br>Fully Plasticized |                 |   |
|---|---|----------------|--|--------------------------------------|----------------|--|---|----------------|--|---|-----------------|---|
|   |   |                |  | Summer Grade                         |                |  | Winter Grade                                    |                |  |   |                 |   |
|   | Min.                                    | Max.           |  | Min.                                 | Max.           |  | Min.  | Max.           |  | Min.  | Max.            |   |
| Softening Point, °C (°F), ASTM D 36               | 99<br>(210)                             | --             |  | 104<br>(220)                         | 116<br>(240)   |  | 104<br>(220)                                    | 116<br>(240)   |  | 116<br>(240)                                    | --              | 127<br>(260)                            |
| Penetration, ASTM D 5                             |   |                |  |                                      |                |  |   |                |  |   |                 |   |
| 25°C (77°F) 100 g/s (3.53 oz/s)                   | 2                                       | 9              |  | 5                                    | 10             |  | 10  | 20             |  | 2   | 6               | 6                                       |
| 46°C (115°F) 50 g/s (1.8 oz/s)                    | 7                                       | 25             |  | 12                                   | 30             |  | 15  | 55             |  | 3   | 15              | 15                                      |
| Ash % by weight, ASTM D 2415                      | 25                                      | 35             |  | 25                                   | 35             |  | 25  | 35             |  | 25  | 35              | 35                                      |
| Specific Gravity at 25°C (77°F), ASTM D 71        | 1.4                                     | 1.6            |  | 1.4                                  | 1.6            |  | 1.4   | 1.6            |  | 1.4   | 1.6             | 1.6                                     |
| High-Temperature Sag Test, cm (in.), AWWA C 203   | --                                      | 0.15<br>(0.06) |  | --                                   | --             |  | --  | --             |  | --  | --              | --                                      |
| 5 h at 66°C (150°F)                               |   |                |  |                                      |                |  |   |                |  |   |                 |   |
| High-Temperature Sag Test, cm (i.), AWWA C 203    | --                                      | --             |  | --                                   | 0.15<br>(0.06) |  | --  | 0.15<br>(0.06) |  | --  | --              | --                                      |
| 24 h at 71°C (160°F)                              |   |                |  |                                      |                |  |   |                |  |   |                 |   |
| High-Temperature Sag Test, cm (i.), AWWA C 203    | --                                      | --             |  | --                                   | --             |  | --  | --             |  | --  | 0.16<br>(0.063) | (at 93°C<br>[200°F])<br>0.16<br>(0.063) |
| 24 h at 82°C (180°F)                              |   |                |  |                                      |                |  |   |                |  |   |                 |   |
| Low-Temperature Crack Test, AWWA C 203            |   |                |  |                                      |                |  |   |                |  |   |                 |   |
| 5 h at -18°C (0°C)                                | None                                    | None           |  | --                                   | --             |  | --  | --             |  | None  | None            | None                                    |
| 6 h at -23.3°C (-10°F)                            | --                                      | --             |  | None                                 | None           |  | --  | --             |  | --  | --              | --                                      |
| 6 h at -29°C (-20°F)                              | --                                      | --             |  | --                                   | --             |  | None  | None           |  | --  | --              | --                                      |
| Peel Test at 27°C (80°F); 71°C (160°F) AWWA C 203 | None                                    | None           |  | None                                 | None           |  | None  | None           |  | None  | None            | None                                    |

Table II

To form a tar-based enamel according to the invention a mixture is formed containing a filler and the enamel base described above containing bases derived from both bituminous and petroleum sources. The mixture heated and mixed at a temperature at least about 225°C to maintain the enamel base as a fluid melt and facilitate mixing. Heating and mixing are continued until the mixture has a uniform composition. Tar-based enamels meeting product specifications can be recovered using conventional means.

As discussed above, the present invention provides metal articles having all or at least a portion of least one surface coated with applicant's novel tar-based enamel. The improved coated metal articles are waterproofed and protected against corrosion. The more preferred coated articles include coated steel pipes and coated steel tanks. The tar-based enamels can be applied by conventional means at elevated temperatures such as for example, spraying, brushing, dipping or flooding, and can be used alone or over an epoxy primer as described in U.S. patent 5,567,480.

For application of a tar-based enamel to a metal article, the enamel is preferably heated to a temperature of about 200°C to about 300°C, more typically in the range of about 230°C to about 280°C. At these temperatures, the tar-based enamel is flowable and can be applied to the metal article. In typical applications, the coal tar enamel will be applied to provide a relatively thick coating, for example in the range of about 90 to about 160 mils. Coatings of such thickness provide serviceable coatings without the presence of polymerizable materials such as epoxies in the topcoat. The tar-based enamels used in the

invention also form a hardened coating relatively quickly, for example, usually within a period of twenty-four hours or less. As with conventional coal tar enamels, it may be advantageous before the tar-based enamel has hardened to reinforce the coating with fiberglass and/or Kraft paper wrapping.

Depending on the article and its use, the coating can be applied to either an exterior or an interior surface. For example, the tar-based enamel coating is generally applied to an exterior surface of a steel pipe, whereas the enamel can be applied to an interior surface, an exterior surface or to both surfaces of a steel tank depending on its service. Interior coatings can utilize tar-based enamels with lower levels of filler and can even utilize an enamel base having a softening point of at least about 90°C without a filler for some limited applications where the coating's impact resistance and sufficient mechanical strength is not important.

Because applicant's tar-based enamels contain reduced levels of PAH's workers coating and installing the coated metal articles as well as the surrounding environment are exposed to lower PAH levels. Applicant's preferred tar-based enamel coatings having sufficiently reduced PAH levels will be suitable for coating the interior of metal tanks containing potable water.

For the purposes of providing a further understanding of the invention and its preferred features and advantages, the following specific examples are provided. It will be understood, however, that these examples are illustrative and not limiting of the invention. The softening points and penetrations reported in the examples provided were carried out according to ASTM methods D36 and D5, respectively.

### Examples 1-8, Enamel Bases from Coal Tar

A mixture of coal tar, CD 103 petroleum oil, slurry oil, and coal dust was slowly heated with stirring. Distillate was removed until the liquid temperature reached about 310 to 320°C where it was maintained without further removal of distillate. After about 30 minutes samples of the hot melt were taken and softening points and penetrations were determined. The melt was heated further to raise the liquid temperature in increments of about 20 to about 40°C and additional distillate was removed. The steps of maintaining the liquid temperature without removal of additional distillate were repeated to allow for further sampling and testing. When the desired softening points and penetrations were obtained the removal of distillate was stopped. The removal of distillate can also be carried out at pressures less than ambient pressures provided a liquid temperature of at least about 300°C is attained.

| Example No.         | 1    | 2    | 3    | 4    | 5    | 6    | 7    | 8    |
|---------------------|------|------|------|------|------|------|------|------|
| Wt.% Coal Tar       | 35   | 35   | 35   | 35   | 35   | 35   | 35   | 35   |
| Wt.% CD-103         | 33.2 | 33.1 | 33   | 33.2 | 33.2 | 33   | 32.5 | 32   |
| Wt.% Slurry Oil     | 15   | 14.9 | 14.5 | 13.8 | 13.3 | 15   | 15   | 15   |
| Wt.% Coal Dust      | 16.8 | 17   | 17.5 | 18   | 18.5 | 17   | 17.5 | 18   |
| Total Blend, g      | 300  | 300  | 300  | 300  | 300  | 300  | 300  | 300  |
| Distilled to °C     | 380  | 350  | 350  | 350  | 340  | 350  | 350  | 350  |
| Softening Point, °C | 67.7 | 62   | 52   | 108  | 110  | 85   | 71.5 | 71   |
| Penetration @77°F   | 18   | 37   | 36   | 17   | 16   | 11   | 24   | 17   |
| %Yield              | 87   | 87.8 | 88.2 | 86.7 | 86.7 | 88.9 | 89.6 | 87.2 |

Table III

### Examples 9-13, Tar-Based Enamels Derived from Coal Tar

The enamel base was prepared by the method described above (Examples 1-8) and brought to a liquid temperature of about 150°C, stirring was initiated and the liquid temperature raised to maximum temperature of about 250°C. For Examples 9, 10, 12 and 13, talc was slowly added with continued heating and stirring and when addition was complete the blend was stirred for an additional 30 minutes at about 250°C. Samples were removed and their softening points and penetrations determined.

| Example No.         | 9     | 10   | 11   | 12   | 13   |
|---------------------|-------|------|------|------|------|
| Wt.% Coal Tar       | 35    | 35   | 35   | 35   | 20   |
| Wt.% CD-103         | 31.5  | 33.2 | 32.7 | 33.5 | 35   |
| Wt.% Slurry Oil     | 15    | 13.3 | 13.8 | 12.5 | 25   |
| Wt.% Coal Dust      | 18.5  | 18.5 | 18.5 | 19   | 20   |
| Total Blend, g      | 300   | 3500 | 3500 | 3500 | 3500 |
| Distilled to °C     | 350   | 355  | 350  | 340  | 350  |
| Softening Point, °C | 103.5 | 108  | 99   | 95   | 105  |
| Penetration @77°F   | 16    | 11   | 13   | 11   | 11   |
| %Yield, enamel base | 87.1  | 91.7 | 94.5 | 94.5 | 84.8 |
| Added Talc (%)      | 30    | 30   | None | 30   | 30   |
| Softening Point, °C | 110.4 | 116  |      | 120  | 107  |
| Penetration @77°F   | 10    | 10   |      | 10   | 7    |

Table IV

Similar tar-based enamels were prepared substituting slate, mica, limestone, silica and kalon for the talc.

A more complete determination of the properties of the tar-based enamel from Example 10 was carried out and is provided below in Table V.

| Physical Property   | Results               |
|---|-----------------------|
| Softening Point, °C (°F), ASTM D 36                           | 116°                  |
| Penetration, ASTM D 5   |                       |
| 25°C (77°F) 100 g/s (3.53 oz/s), 1/10 mm                      | 10                    |
| Ash % by weight, ASTM D 2415                                  | 30                    |
| Specific Gravity at 25°C (77°F), ASTM D 71                    | 1.5                   |
| High-Temperature Sag Test, cm (in.), ANSI/AWWA C 203          |                       |
| 24 h at 71°C (160°F)  | < 0.15 cm (< 0.06 in) |
| Impact Test, @ 25°C (77°F), 650 g ball, 8 ft drop, AWWA C 203 |                       |
| Direct Impact, disbonded area                                 | 3.4 in <sup>2</sup>   |
| Low-Temperature Crack Test, ANSI/AWWA C 203                   |                       |
| 6 h at -23.3°C (-10°F)  | None                  |

Table V

Examples 14-22, Tar-Based Enamels Derived from Refined Coal Tar - Driveway Sealer Tar (DST Tar)

(a) Preparation of Enamel Base: A mixture of DST grade refined coal tar, CD 103 clarified catalytic cracker oil, slurry oil, and coal dust was slowly heated with stirring. Distillate was removed until the liquid temperature reached about 305°C where it was maintained for about one hour without further removal of distillate. While maintaining the melt at about 300°C samples were taken and softening points and penetrations determined.

(b) Preparation of Tar-Based Enamels: The enamel base was prepared by the method described above and brought to a liquid temperature of about 150°C, stirring was continued and the liquid temperature raised to maximum temperature of about 250°C. Talc was slowly added with continued heating and stirring and

when addition was complete the blend was stirred for an additional 30 minutes at about 250°C. Samples were removed and their softening points and penetrations determined. Similar tar-based enamels were prepared substituting slate, mica, limestone, silica and kalon for the talc.

| Example No.         | 14    | 15    | 16    | 17    | 18    | 19    | 20    | 21    | 22    |
|---------------------|-------|-------|-------|-------|-------|-------|-------|-------|-------|
| Wt.% DST Tar        | 34.4  | 35.8  | 34.0  | 34.3  | 34.4  | 34.0  | 34.1  | 34.0  | 33.9  |
| Wt.% CD-103         | 25.5  | 24.4  | 23.0  | 25.5  | 25.4  | 25.0  | 25.4  | 24.0  | 24.0  |
| Wt.% Slurry Oil     | 18.5  | 18.4  | 21.5  | 19.2  | 19.7  | 22.0  | 22.0  | 23.5  | 24.0  |
| Wt.% Coal Dust      | 21.6  | 21.4  | 21.5  | 21.0  | 20.5  | 19.0  | 18.5  | 18.5  | 18.5  |
| Total Blend, g      | 601.9 | 603.9 | 601.3 | 602.5 | 600.0 | 600.4 | 601.3 | 600.2 | 600.0 |
| Distilled to °C     | 305   | 305   | 305   | 300   | 305   | 305   | 305   | 307   | 305   |
| Softening Point, °C | 159   | 156   | 149   | 179   | 172   | 132   | 125   | 127   | 125   |
| Penetration @77°F   | 11    | 8     | 6     | 8     | 10    | 15    | 37    | 41    | 21    |
| %Yield, Enamel Base | 97.7  | 97.7  | 99.0  | 98.6  | 98.0  | 98.2  | 99.0  | 99.0  | 99.0  |
| Added Talc (%)      | 20    | 20    | 20    | 20    | 20    | 20    | 20    | 20    | 20    |
| Softening Point, °C | 159   | 156   | 143   | 176   | 175   | 149   | 135   | 132   | 125   |
| Penetration @77°F   | 7     | 8     | 6     | 6     | 10    | 10    | 19    | 12    | 18    |

Table VI

#### Examples 23-30, Enamel Bases from Coal Tar Pitch

A mixture of coal tar pitch, CD 103 clarified catalytic cracker oil, slurry oil, and coal dust was slowly heated without stirring to about 100-120°C to create a melt that could be agitated. Stirring was initiated and the melt was further heated to a liquid temperature of about 320°C with the removal of distillate. While maintaining the melt at about 320°C without the removal of additional distillate samples were taken and softening points and penetrations determined. The melt

was heated further to raise the liquid temperature in increments of about 10 to 15°C and additional distillate was removed. The steps of maintaining the liquid temperature without removal of additional distillate was repeated to allow for further sampling and testing. The softening points and penetrations were again determined. When the desired softening points and penetrations were obtained the removal of distillate was stopped.

| Example No.         | 23   | 24   | 25   | 26   | 27   | 28   | 29   | 30   |
|---------------------|------|------|------|------|------|------|------|------|
| Wt.% 60°C Pitch     | 20   | 20   | 20   | 20   | 22   | 20   | 20   | 20   |
| Wt.% CD-103         | 20   | 20   | 23   | 27   | 28   | 24   | 20   | 22   |
| Wt.% Slurry Oil     | 41   | 40   | 36.5 | 32.5 | 27.5 | 36   | 40.5 | 38   |
| Wt.% Coal Dust      | 19   | 20   | 20.5 | 20.5 | 22.5 | 20   | 19.5 | 20   |
| Total Blend, g      | 300  | 300  | 300  | 300  | 300  | 300  | 300  | 300  |
| Distilled to °C     | 335  | 350  | 335  | 335  | 340  | 345  | 355  | 360  |
| Softening Point, °C | 98   | 88   | 85   | 68   | 82   | 79   | 98   | 98   |
| Penetration @77°F   | 6    | 6    | 6    | 6    | 5    | 9    | 4    | 4    |
| %Yield, Enamel Base | 90.5 | 92.1 | 92   | 92.5 | 89.3 | 89.4 | 85.6 | 86.5 |

Table VII

#### Examples 31-35, Tar-Based Enamels Derived from Coal Tar Pitch

The enamel base was prepared by the method described above (Examples 23-30) and brought to a liquid temperature of about 150°C, stirring was initiated and the liquid temperature raised to maximum temperature of about 250°C. Talc was slowly added with continued heating and stirring and when addition was complete the blend was stirred for an additional 30 minutes at about 250°C. Samples were removed and their softening points and penetrations determined.

| Example No.         | 31   | 32    | 33  | 34   | 35   |
|---------------------|------|-------|-----|------|------|
| Wt.% 60°C Pitch     | 20   | 20    | 20  | 20   | 20   |
| Wt.% CD-103         | 23   | 23    | 22  | 22.5 | 23   |
| Wt.% Slurry Oil     | 37   | 37    | 37  | 37.5 | 38   |
| Wt.% Coal Dust      | 20   | 20    | 21  | 20   | 19   |
| Total Blend, g      | 300  | 4,000 | 600 | 601  | 600  |
| Distilled to °C     | 350  | 320   | 310 | 310  | 307  |
| Softening Point, °C | 101  | 86    | 168 | 148  | 125  |
| Penetration @77°F   | 6    | 9     | 10  | 15   | 41   |
| %Yield, Enamel Base | 87.4 | 88    | 95  | 97   | 98.9 |
| Added Talc (%)      | 20   | 30    | 20  | 20   | 20   |
| Softening Point, °C | 145  | 112   | 168 | 167  | 166  |
| Penetration @77°F   | 3    | 7     | 10  | 15   | 15   |

Table VIII

The PAH content of a typical coal tar enamel prepared from 100% bituminous bases, and several hybrid tar-based enamels and enamel bases were examined to determine their total B(a)P equivalent level and the percent reduction for the tar-based enamels and enamel bases. Typical results are provided below in Table IX.

|                              | Total B(a)P | % Reduction |
|------------------------------|-------------|-------------|
| Typical Coal Tar Enamel      | 12167.92    | --          |
| Enamel Base, Example 13      | 7245.10     | 43.78%      |
| Tar-Based Enamel, Example 13 | 6883.76     | 46.97%      |
| Tar-Based Enamel, Example 10 | 10,166.70   | 21.11%      |

Table IX

All publications and patents cited herein are hereby incorporated by reference in their entirety as if each had been individually incorporated by reference and fully set forth.

While the invention has been described in some detail in the foregoing passages, the same is to be considered as illustrative and not restrictive in character, it being understood that only the preferred embodiments have been described and that all changes and modifications that come within the spirit of the invention are desired to be protected.